SEMICLASSICAL STUECKELBERG OSCILLATIONS OF THE MICROWAVE-DRIVEN EXCITED HYDROGEN ATOM

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ABSTRACT. First experiments are described that probe coupled pairs of quantum quasienergy eigenstates of highly excited hydrogen atoms in strong pulsed microwave fields. During the rise and fall of the pulse, initial states located near a classical nonlinear resonance region of the microwave frequency are found to couple to final states with a very different zero-field principal quantum number. Oscillations in the ionization probability as a function of pulse peak field are observed to satisfy classical scaling laws, revealing semiclassical characteristics of the coupled two-state system.

Experiments on highly excited hydrogen atoms in microwave fields have proven helpful in the development of the theory of nonintegrable semiclassical quantum systems possessing interesting underlying classical nonlinear dynamics. In particular, the latter can be chaotic, leading to ionization of the atom [1]. This can occur when the initial Kepler electron orbiting frequency, the microwave frequency and the quantum two-state resonant Rabi frequency all are comparable in magnitude. This time-periodic strong field regime quantum system can be addressed using Floquet theory, which introduces the unitary time evolution operator for one microwave period of time, the related quasienergy operator, and the latter's quasienergy eigenvalues and eigenstates [2,3]. The quasienergies play the role of the quantized energies of a conservative system, and are functions of the fixed microwave field strength F.

In experiments the microwave field actually is pulsed, so that the adiabatic quasienergy state adiabatically connected to the initial free-atom state just follows the rise and fall of the pulse. However, calculations show that quasienergy level crossings and avoided crossings abound in field plots of the quasienergies [3]. In the lower-F portion of the semiclassical regime, the classical dynamics is regular and the quasienergy nearest neighbor separation distribution is Poisson; here the percentage of true crossings is relatively large [4]. However, at strongly ionizing ("chaotic") higher values of F the distribution becomes Wigner and all crossings are significantly avoided. In the experimental pulsed optical ionization of ground state atomic hydrogen, there is some evidence that some results are described by the evolution of a single diabatic quasienergy state [5]. For microwave-driven hydrogen, some experimental results appear connected with primarily one quasienergy state being populated at the peak of the microwave pulse [6].

There are predictions that quasienergy level crossing effects should be important for atomic and molecular processes induced by strong short-pulse electromagnetic fields [7]. However, there has been little experimental evidence outside of perturbative regimes. This could be due to spatial

field nonuniformity in strong field optical experiments, and to initial atomic state averaging in most past microwave-driven hydrogen experiments. We employ experimental techniques that simultaneously address both these problems. In this paper we present the first observations of large quasienergy level crossing effects for the microwave-driven hydrogen system.

Stueckelberg oscillations arise through a parameter dependence in an interference of amplitudes of two quantum states that are sequentially mixed at two well separated localized times [8]. Such oscillations are a well-known phenomenon in atom-atom collisions, where the localized state-mixing is near a pseudocrossing of potential energy curves and the time evolution through the pseudocrossing is driven by the changing atom-atom separation. These oscillations also have been observed recently in pulsed microwave multiphoton excitation experiments using K and He Rydberg atoms [9]. These experiments were in a weak field strength regime, where multiphoton perturbation theory can be used to compute both diabatic quasienergies and their couplings as a function of field strength. One quasienergy state was initially populated and a coupled second quasienergy state was detected by auxiliary static electric field ionization. The present experiments on single-state highly excited hydrogen atoms differ in that the peak strength of the driving microwave field typically is much higher, in a regime where the probability of direct microwave "ionization" can be above 1% and many but not necessarily all features of the atom-field system are semiclassical [6].

Our experiments utilized the well-known optical double resonance fast atomic beam technique [10,11]. A proton beam from a small accelerator is converted into a mixed-state hydrogen beam by electron transfer collisions in xenon gas, the remaining protons being subsequently electrostatically deflected from the beam. Then all atoms in the beam with principal quantum number $n > 9$ are removed by field ionization in a strong electric field. The remaining mixed-state atom beam then traverses two laser excitation regions in turn, each determined by an appropriate

static electric field. In the first region, those atoms in the beam by happenstance being in the lowest energy or "stretched atom" state of the $n = 7$ Stark state manifold are resonantly excited by a CW carbon dioxide laser beam to the analogous state with $n = 10$. In the second region a similar process further stretches the atom along the static field direction, producing atoms with parabolic quantum numbers $n, n_1, m = n_o, 0, 0$, where n_o can be selected to be either 59, 60 or 61 through adjustment of the static field, the laser line and the Doppler shift due to atom kinetic energy. In our approach, the second static field F_s is a motional field $v \times B$ [10]. The atoms then pass through circular holes in the sidewalls of a copper TE10 rectangular waveguide, where F_s is still present. In an atom's reference frame, it is exposed to a pulse of microwave electric field having a half sinewave pulse shape and a pulse length of about 100 microwave periods, the exact value depending on the microwave frequency. All static electric fields as well as the microwave electric field are collinear to within a few milliradians, maintaining stretched atom states during the atom preparation, microwave interaction, and atom detection stages of the experiment [10].

Microwave "ionization" was experimentally defined as true ionization plus excitation to quantum numbers outside the presently maximal range $n_b = 50(3)$ to $n_c = 90(3)$. Atoms within this range were detected with at least 95% efficiency using field ionization. For various values of microwave frequency ω between 12.4 and 18.0 GHz, of static field strength F_s within the microwave interaction region between 6.0 and 10.0 V/cm, and of residual r.m.s. microwave noise field between 0.2% and 5%, analog recordings were made of the ionization probability P_I as a function of r.m.s. waveguide microwave power. Several recordings generally taken on different days were averaged for each set of parameters, and our error bars indicate the spread in values over the recordings. The possibility of any laser excitation of atoms within the microwave region was monitored by recordings taken with the atoms preionized away in a field before the microwave region; such excitation was never observed and could amount to at most 3% probability. Our studies of the effects of additional extrinsic microwave noise, when linearly

extrapolated to zero noise power, indicate that the present ionization data are not altered by such noise to within 5% probability. A check was made for effects arising from microwave harmonics by removing the microwave amplifier and reproducing some low-field data.

Following tradition, we define the ionization threshold value F(10%) of peak microwave electric field strength to be that for 10% ionization probability. The dependence of the ionization threshold on microwave frequency was found to exhibit unexpected features, see figure 1. Here F and ω have been classically scaled to characteristic values for the initial state of the atom and furthermore ω has been corrected for the value of F_s, according to $F_o = n_o^4 F$ and $\omega_o' = n_o^3\omega/(1-3n_o^4 F_s)$ [6]. The six features displaying ionization threshold reduction exhibit linear increases with decreasing microwave frequency, according to $F_o(10\%) = C[\omega_o'(0)-\omega_o']$ with C = 2.05(0.25) and $\omega_o'(0)$ = 0.520, 0.537, 0.558, 0.576(0.003) for features 3 through 6 respectively. Their adjacent feature spacing is 0.0185(0.002).

That the origin of the ionization threshold reduction is coupling of the initial quantum state to a second quantum state is clear from the Stueckelberg oscillations in the ionization probability as a function of peak microwave field strength or of microwave waveguide power, see figure 2. Each feature in figure 1 has an appearance threshold, defined as the highest frequency where an oscillation still reaches 10% ionization probability. At and somewhat below such frequencies, only one maximum in P_I was observable. At still lower ω_o', additional maxima appeared above the first one, but none below it. Hence the first maximum corresponds to the peak pulse field strength reaching a level-crossing value F_c. We also note that the spacing in F_o of adjacent maxima was very different for each feature, indicating coupling of different quasienergy states.

We investigated the role of the static electric field F_s in our system. A 20% change in F_s will produce about a 1% change in ω_o'. The resultant changes in the entire oscillation pattern were easily observed and experimentally could be compensated for by readjusting the microwave frequency ω to keep ω_o' fixed. Therefore the magnitude of the static field is not critical for the existence of the oscillations.

A test for classical scaling involves changing the initial principal quantum number n_o of the atom while keeping the scaled microwave field F_o and scaled microwave frequency ω_o' fixed. Without fixing these, a 1.6% change in n_o produced changes of about 5% and 6.5% in ω_o' and F_o respectively, which resulted in completely different ionization probability curves. Only when we compensate by adjusting both ω and F to satisfy classical scaling do we get very similar oscillations for different values of n_o, as shown in figure 3. The onsets and spacings are in close agreement. This shows that differences in quasienergies of the involved quasienergy states can satisfy a classical scaling, and is strong evidence for semiclassical characteristics of coupled pairs of strong-field quasienergy states. The observed scaling is consistent with past experimental observations of scaling of ionization probability [14]. Theory suggests a second scaling for individual quasienergy states, that however does not concern level separations [15].

Previous numerical calculations of the field dependence of the quasienergies (with $F_s = 0$) indicate correspondences with classical features in sectioned phase space [3,7,13,15]. In brief, although at sufficiently low F_o the quasienergies decrease quadratically with increasing F_o, at strong fields the behavior is almost linear. In the $\omega_o' = 1/2$ nonlinear resonance region, the slope is weakly negative, in the $\omega_o' = 1$ resonance region it is strongly positive, and it is strongly negative both between these regions and just beyond these regions. However, modifications due to $F_s > 0$ have not been investigated.

We consider an initial quasienergy state Ψ_1 near the $\omega_o' = 1/2$ nonlinear resonance region and some final state Ψ_2. We model the strong-field diabatic quasienergies by $E_1(F_o) = E_1(0) + a_1F_o$ and

$E_2(F_o) = E_2(0) + a_2F_o$. The level crossing condition $E_2 - E_1 = N\omega$ for some integer N gives an equation for the crossing field strength $F_c = [E_2(0)–E_1(0)–N\omega]/(a_1–a_2) = A–B\omega_o$. Comparing this with the experimental formula for F(10%) quoted above, the sign of C requires that A and B be positive, and the signs of $E_2(0) - E_1(0)$ and N are the same. Thus there are two possibilities for

the states Ψ_2 : 1) $N > 0$ with $n > n_c– 3 = 87$, $N > 32$ and $a_1–a_2 > 0$, or 2) $N < 0$ with $n < n_b+3 =53$, $–N > 16$ and $a_1–a_2 < 0$. The first possibility places $E_2(0)$ at or above $\omega_o' = 3/2$, and the second at or below $\omega_o' = 1/3$, where a_2 is not known. Thus identification of the various states Ψ_2 awaits further quasienergy calculations and experimental improvements in the ranges of n_b and n_c.

The Landau-Zener-Stueckelberg (LZS) formula is derived for P_I using standard techniques for constructing the Landau-Zener diabatic time evolution matrices through sharp crossing regions [16] and a matrix for adiabatic evolution between the crossings. Given the present experimental relationship between time and instantaneous r.m.s. field strength, the Landau-Zener parameter is $\delta = V_o^2T/\{\alpha F_c[(F_m^2/F_c^2–1)]^{1/2}\}$ and determines a diabatic phase change $\phi'(\delta)$ [16]. Here V_o is the separation of the adiabatic quasienergies at F_c, F_m is the pulse peak field, α is proportional to the difference in slopes of the modeled diabatic quasienergies at F_c and T is the pulse duration. P_I is given by $4e^{-2\pi\delta}(1-e^{-2\pi\delta})\sin^2(\phi-\phi')$, where ϕ is the phase difference between Ψ_1 and Ψ_2 accumulated between the two crossings. However, the oscillations are usually observed to become damped at higher F_m, as expected from the presence of further important avoided crossings to additional states. We model this damping by including (exponential) loss factors during the evolution between the primary level crossings. Then we obtain

$$P_I = (1-e^{-2\pi\delta})(1-e^{-\gamma_2}) + e^{-2\pi\delta}(1-e^{-\gamma_1})$$
$$+e^{-2\pi\delta}(1-e^{-2\pi\delta})[(e^{-\gamma_1/2} - e^{-\gamma_2/2})^2 + 4e^{-(\gamma_1+\gamma_2)/2}\sin^2(\phi-\phi')] \quad , \qquad (1)$$

where the first two terms represent loss between the crossings. We have carried out multiparameter least squares fits of our data to this expression. For all the data, a reasonable fit can be found, see figure 2 for an example. The values obtained for F_c and α are robust in that they

are independent of existence assumptions about the loss exponents γ_1, γ_2, to within ±10%. This is because F_c and α are determined primarily by the onset and spacings of the Stueckelberg oscillations.

We have found that coupled quasienergy states can produce classically scalable features in transition probabilities, indicating that the latter have semiclassical characteristics. However, further experiments up to $\omega_o' = 1.2$ have not revealed any more Stueckelberg oscillations, for the selected cutoff quantum numbers n_b and n_c. The Husimi quantum electron probability distributions of the quasienergy states, in sectioned phase space, are expected to exhibit features associated with specific types of classical electron trajectories [6,17]. Such distributions might help clarify why the Stueckelberg oscillations have appeared only for scaled microwave frequencies in the $\omega_o' = 1/2$ nonlinear resonance region.

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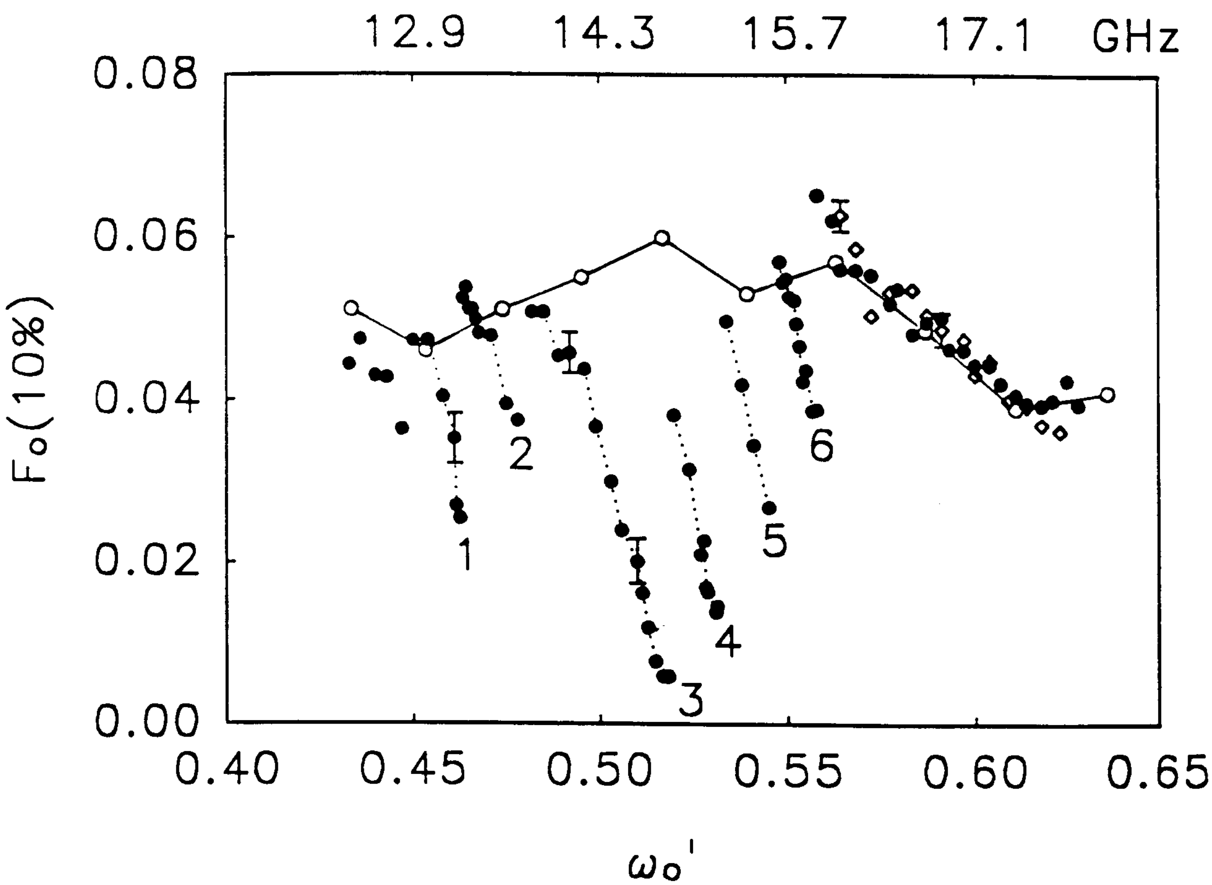


FIGURE 1. Dependence of the scaled microwave ionization threshold field strength upon the Stark-corrected scaled microwave frequency ω_o'. A comparison of present short-pulse stretched atom data is made to previous long-pulse three-dimensional data (solid line) at zero static field [12]. Dots : n_o = 60, and diamonds : n_o = 65, are present data with a static field F_s = 8.0 V/cm. The upper cutoff quantum number defining "ionization" is n_c = 90. Reduction of the ionization threshold field below expected values is observed for features 1 through 6.

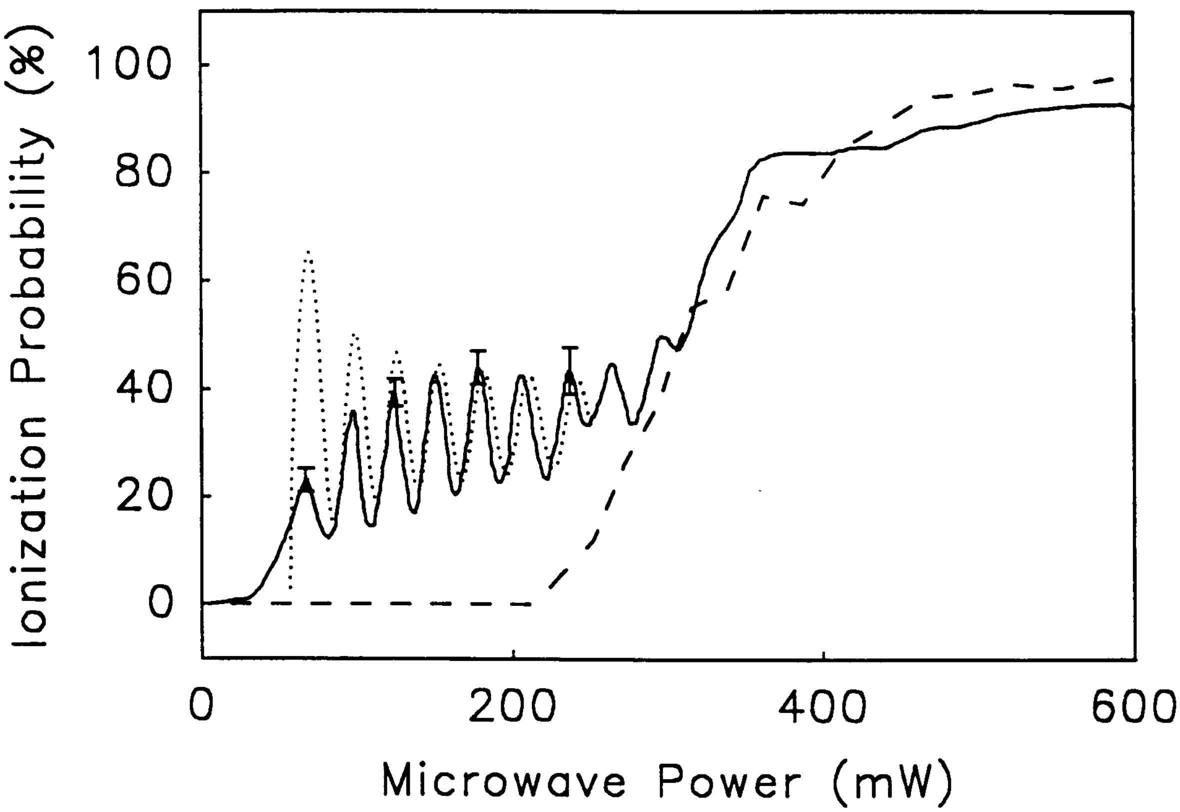
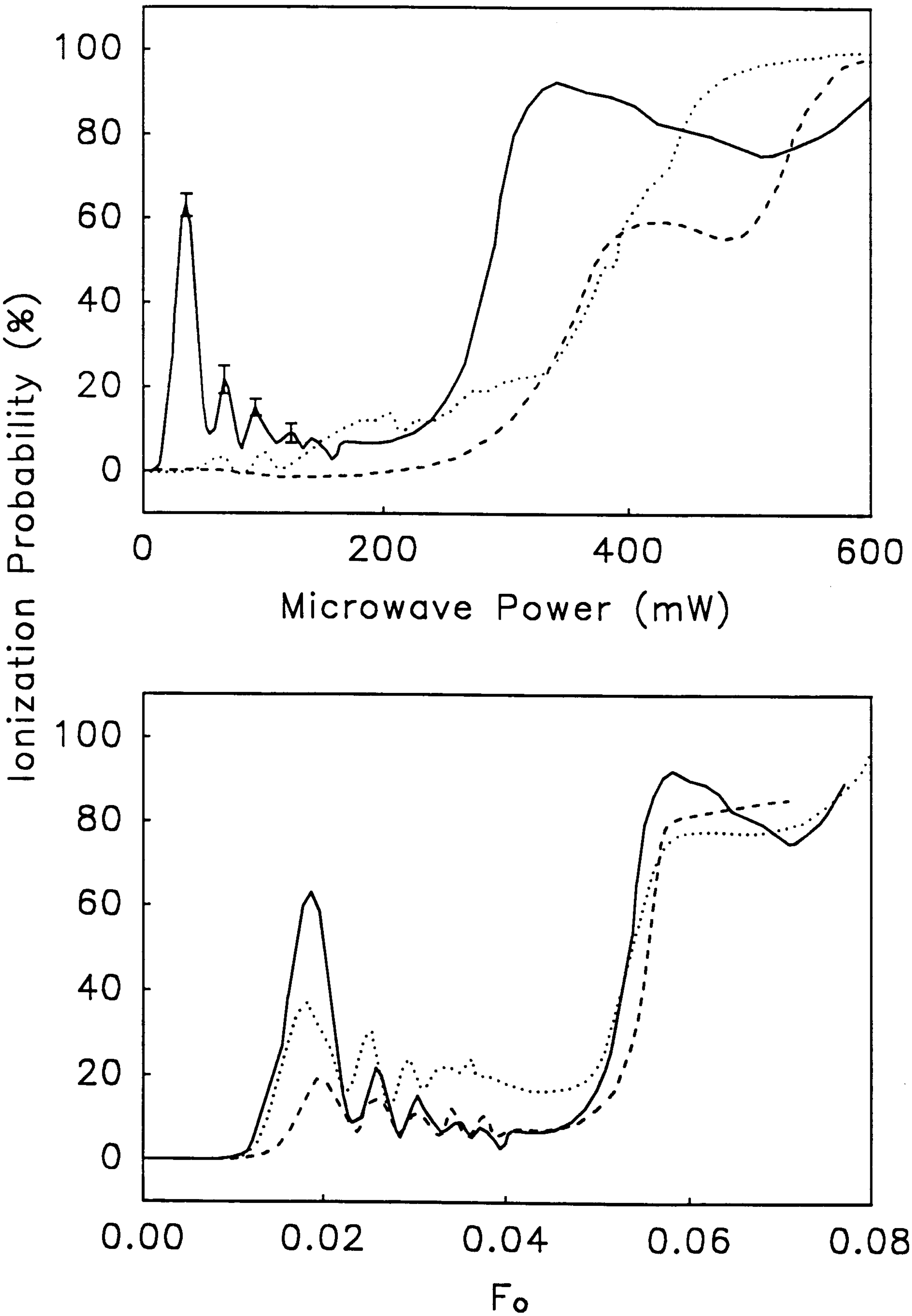


FIGURE 2. Ionization probability versus r.m.s. waveguide microwave power, for n_o = 60 stretched atoms, a microwave frequency of 15.10 GHz, and F_s = 8.0 V/cm. The solid line is experimental, the dashed line the result of one-dimensional classical numerical calculations, and the dotted line the result of a fit to a Landau-Zener-Stueckelberg quantum level crossing model. This is for feature 4 at ω_o' = 0.528.



100
80
60
40
20
0
0
200
400
600
Microwave Power (mW)
Ionization Probability (%)
100
80
60
40
20
0
0.00
0.02
0.04
0.06
0.08
Fo

FIGURE 3. Ionization probabilities for three different atom initial quantum numbers, n_o = 59 (dots), 60 (solid lines) and 61 (dashed lines). Top : Dependence on waveguide microwave power, at a fixed unscaled microwave frequency of 15.20 GHz. The three curves are quite different. Bottom: Dependence on scaled microwave field strength, at $\omega_o' = 0.531$. Feature 4 Stueckelberg oscillations are observed for all three quantum numbers, with the same scaled field onset and maxima spacings.

REFERENCES

1. M. I. Meerson, E. A. Oks and P. V. Sasorov, Pis'ma Zh. Eksp. Teor. Fiz. **29**, 79 (1979) [JETP Lett. **29**, 72 (1979)].

2. H. Sambe, Phys. Rev. A **7**, 2203 (1973).

3. H. P. Breuer, K. Dietz and M. Holthaus, Z. Phys. D **10**, 13 (1988); Z. Phys. D **11**, 1 (1989).

4. R. Blumel and U. Smilansky, Zeit. Phys. D **9**, 95 (1988).

5. M. Dorr, D. Feldman, R. M. Potvliege, H. Rottke, R. Shakeshaft, K. H. Welge and B. Wolff-Rottke, J. Phys. B **25**, L275 (1992).

6. R. V. Jensen, S. M. Susskind and M. M. Sanders, Phys. Rpts. **201**, 1 (1991).

7. H. P. Breuer and M. Holthaus, Z. Phys. D **8**, 349 (1988).

8. E. C. G. Stueckelberg, Helv. Phys. Acta **5**, 369 (1932); W. R. Thorson, J. B. Delos and S. A. Boorstein, Phys. Rev. A **4**, 1052 (1971).

9. M. C. Baruch and T. F.Gallagher, Phys. Rev. Lett. **68**, 3515 (1992); S. Yoakum, L. Sirko and P. M. Koch, Phys. Rev. Lett. **69**, 1919 (1992).

10. J. E. Bayfield and L. A. Pinnaduwage, Phys. Rev. Lett. **54**, 313 (1985); J. E. Bayfield, in *Quantum Measurement and Chaos*, E. R. Pike and S. Sarkar, editors, Plenum Press, 1987, pp 1-33; J. E. Bayfield and D. W. Sokol, Phys. Rev. Lett. **61**, 2007 (1988).

11. P. M. Koch, in *Rydberg States of Atoms and Molecules*, R. F. Stebbings and F. B. Dunning, editors. Cambridge University Press, 1983, pp 473-512.

12. M. M. Sanders, R.V. Jensen, P. M. Koch and K. A. H. van Leeuwen, Nuc. Phys. B (Proc. Suppl.) **2**, 578 (1987).

13. H. P. Breuer and M. Holthaus, J. Phys. II **1**, 437 (1991).

14. J. E. Bayfield, D. W. Sokol, G. Casati and I. Guarneri, Phys. Rev. Lett. **63**, 364 (1989).

15. H. P. Breuer and M. Holthaus, Ann. Physics **211**, 249 (1991).

16. J. E. Bayfield, E. E. Nikitin and A. I. Resnikov, Chem. Phys. Lett. **19**, 471 (1973).

17. D. Delande, in *Chaos and Quantum Physics*, A. Voros, J. Zinn-Justin and M. Gianonni, editors. North-Holland, 1990, pp 665-726.